# ROLE OF OXYGEN DIFFUSION IN SELF-HEATING OF FOREST PRODUCTS<sup>1</sup>

## Hans Kubler

Associate Professor
Department of Forestry, University of Wisconsin, Madison, WI 53706
(Received October 1982)

#### **ABSTRACT**

This study deals with self-heating of forest products from oxygen diffusing into the material ('diffusion oxygen'') to be consumed and generate heat. Some or all of this heat is lost through conduction and—in moist material—through diffusing vapor. When self-heating commences, the heating potential of "diffusion oxygen" exceeds heat loss, but since losses increase with temperature more rapidly than does diffusion of oxygen, the materials reach a stable temperature at which all heat that can be generated by oxygen is lost. At 20 C ambient temperature, this theoretical stable temperature is below 25 C in moist solid wood under side grain surfaces, below 40 C under end grain surfaces, but is on the order of 80 to 100 C in piles of moist chips and sawdust, as well as in stacks of fiber insulating board.

At low temperatures, heat lost by conduction exceeds heat transferred by diffusing vapor, even in fiber insulating board. Therefore the stable temperatures for dry solid wood are not much higher than for moist solid wood. Above 50 C, however, heat lost through vapor approaches heat lost through conduction. Therefore, the stable temperatures for permeable materials such as sawdust are much higher in the dry state than in the moist state.

Diffusion of oxygen, heat conduction, and vapor diffusion play a role in the self-heating of all forest products and are decisive for the course of self-heating, not only in the relatively impermeable products such as solid wood, but also in chips, sawdust, bark, and insulating board that are confined by impermeable walls. In open piles of these permeable materials, however, convection of hot air as in a chimney transfers additional heat out of the material. Therefore temperatures in piles of moist chips and sawdust rise to less than between 80 and 100 C, another reason being that not all oxygen entering the piles is consumed.

Keywords: Oxygen diffusion, temperatures, heat conduction.

### INTRODUCTION

Self-heating involves decomposition and devalues forest products in many ways. The products self-heat in piles of chips—particularly chips obtained from whole trees—in piles of sawdust and bark, in hot-stacked fiberboard, in hardboard during the tempering process, in high-temperature dryers, and in structures close to hot items. Oxygen has no part in some of the processes that cause self-heating, namely in the metabolism of anaerobic bacteria, in hydrolysis, and in pyrolysis (Kubler 1982a), but oxygen-consuming microorganisms can raise wood temperatures to the levels of heat required for hydrolysis and pyrolysis. Other self-heating processes depend entirely on oxygen—such as respiration of living wood cells, metabolism of wood-inhabiting fungi, and of course direct chemical oxidation. Without oxygen, wood cannot ignite spontaneously during the final phase of self-heating.

With regard to control of self-heating, it is of interest how the gas gets inside

<sup>&</sup>lt;sup>1</sup> Research supported by the Graduate School and the School of Natural Resources, College of Agriculture and Life Sciences, University of Wisconsin–Madison.

forest products. Some oxygen already fills cell cavities and spaces between pieces before self-heating commences; the relatively small temperatures resulting from consumption of this "residence oxygen" have been calculated earlier (Kubler 1982c). Oxygen is also conveyed into permeable piles by currents of air, driven by the chimney effect in hot piles (Kubler 1982b). The third mode of supply, molecular diffusion, shall be investigated here. It is the objective of this study to determine how much oxygen diffuses into forest products, and how far wood temperatures rise when this oxygen is consumed in wood.

#### APPROACH

Consumption of oxygen inside products creates concentration gradients, which in turn cause diffusion from the outside into the material. Diffusion is slow, but over the long periods of self-heating the amounts of diffusion oxygen add up, and heat released by reaction with oxygen is high (wood's heat of combustion, 20 kJ/g [Hawley 1952], corresponds for wood of the empirical chemical formula  $C_{42}H_{60}O_{28}$  [Klason et al. 1910] to 14.726 kJ per gram of consumed oxygen). The released heat produces a temperature gradient under which the material transfers heat out into the surroundings by means of conduction and, in moist material, of diffusing vapor. The products self-heat to a stable temperature, with generated heat equalling transferred heat. To find this stable temperature, the three types of heat flux are assessed and compared for various temperatures.

Diffusion of oxygen into wood plays no role in fires, where charred wood surfaces react with all the oxygen reaching the surface, leaving none for diffusion into wood. Even if the concentration of oxygen at the surface were higher than inside, the oxygen could hardly diffuse against the pyrolytic gases that stream from the thermally decomposing interior to the surface. Furthermore, combustion times are too short for significant diffusion to occur.

## DIFFUSION OF OXYGEN

Migration of oxygen  $(m_0)$  in direction x (cm) perpendicular to the wood surface is in the steady state, according to Fick's first law,

$$m_o = -D_{oe} \Delta c_o / \Delta x \qquad g/cm^2 s \tag{1}$$

Assuming that concentration of oxygen ( $c_o$ ) at the place of consumption is zero, the differential  $\Delta c_o$  equals the concentration of oxygen in air outside the wood, 0.00028 g/cm<sup>3</sup> at 20 C. The effective diffusivity  $D_{oe}$  depends on temperature T (°K) and is related to the normal molecular or bulk diffusion coefficient  $D_o$  of oxygen in air,  $D_o = 0.178(T/273)^{1.75}$  cm<sup>2</sup>/s (Gray et al. 1972).

In piles of pulp chips and sawdust as well as in fiber insulating board, oxygen diffuses mainly in voids between the pieces, rather than through the pieces. Hence  $D_{oe}$  depends mainly on the fractional voidage, defined as volume of voids per volume of porous pieces of wood (cm³/cm³), and a tortuosity factor in the form  $D_{oe} = D_o V/\tau$  (Coulson and Richardson 1968). The factor  $\tau$  equals the ratio of path length that must be traversed by molecules in diffusing between two points in direction x; it obviously is a function of voidage V, and has to be  $\tau = 1$  for V = 1, and  $\tau = \infty$  for V = 0. The equation  $\tau = 1/V$  meets these requirements, but since path length is related to the volume of the pieces, voidage V should be considered in the third power,  $\tau = 1/V^3$ , so that  $D_{oe} = D_o V^4$ . With V = 0.67 and

V=0.63 for sawdust (indexed s) and chips (c), respectively (Kubler 1982b), we arrive at  $D_{oes}=0.1975D_o$ , and  $D_{oec}=0.1575D_o$ .

At first glance, it seems unlikely that piles of the relatively large chips are less permeable than sawdust. We deal here with diffusion, however, rather than with convection, whose speed due to viscous drag in films at channel surfaces rapidly decreases in narrowing channels. Actually channel width has some influence also on diffusion. Therefore it appears to be reasonable to calculate for sawdust, and for chips with the same effective diffusivity,  $D_{oe} = 0.16D_o$ , so that migration  $m_o$  in piles with voids between pieces becomes

$$m_o = -0.16 \times 0.178(T/273)^{1.75} \times 0.00028/\Delta x$$
  
= -80 × 10<sup>-7</sup>(T/273)<sup>1.75</sup>/\Delta x g/cm<sup>2</sup> s (2)

The amount of heat generated by oxygen, 14,726 J/g, leads to the heat flux  $Q_0$ 

$$Q_0 = -6 \times 10^{-6} \Gamma^{1.75} / \Delta x$$
 J/cm<sup>2</sup> s (3)

In extremely light fiber insulating board of  $10 \text{ lb/ft}^3$  density, made from wood of specific gravity 0.5, the fractional volume of fibers is  $10/(0.5 \times 62.4) = 0.32$ , and the fractional voidage V = 1 - 0.32 = 0.68, or slightly higher than in sawdust. Orientation of the fibers in the plane of the panel restricts diffusion somewhat, so that Eq. 2 can be applied.

Light hardboard of the borderline density 31 lb/ft<sup>3</sup>, lying between insulating board and hardboard, lacks voids between fibers if it is manufactured from a wood species of that same density or specific gravity 0.5. Through this type of panel, gases have to diffuse from cell cavity to cell cavity across cell walls, as in the case of diffusion through solid wood transverse to fiber direction, discussed herewith.

In the absence of experimental data about diffusion of oxygen in cell walls, I assume that cell walls obstruct the passage of oxygen roughly as much as oven-dry cell walls obstruct the passage of water vapor. Accordingly, in solid wood the walls reduce diffusivity  $D_0$  transverse to fiber direction by a factor of 0.01 (Kubler 1957):

$$m_{\text{owt}} = -0.178(T/273)^{1.75} \times 0.01 \times 0.00028/\Delta x$$
 g/cm<sup>2</sup> s (4)

$$Q_{\text{owt}} = -4 \times 10^{-7} \text{T}^{1.75} / \Delta x \qquad \text{J/cm}^2 \text{ s}$$
 (5)

In fiber direction, migration  $(m_{owt})$  and heat flux  $(Q_{owt})$  are about ten times higher:

$$Q_{owf} = -4 \times 10^{-6} \Gamma^{1.75} / \Delta x$$
 J/cm<sup>2</sup> s (6)

The calculated fluxes Q in Figs. 1 to 3 apply to  $\Delta x = 1$  cm, meaning for consumption of oxygen 1 cm below the surface. To obtain flux in case of consumption n cm below the surface, divide the shown values by n.

## HEAT CONDUCTION

Conduction of heat in forest products is complicated by two facts. First, all parts of the unit generate heat, not only its center; accordingly, the temperature gradient  $\partial t/\partial x$  (C/cm) produced by the heat  $Q_o$  should increase towards the sur-

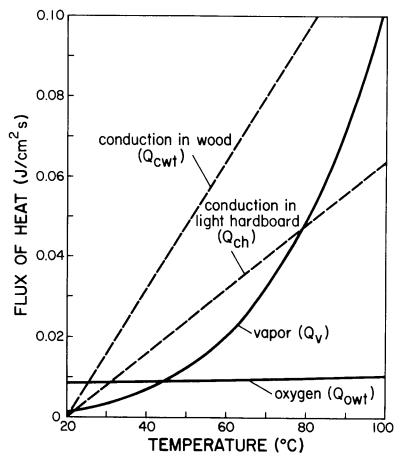


FIG. 1. Transverse fluxes of heat produced by diffusing oxygen, conduction, and diffusing vapor in solid wood and light hardboard at 30% moisture content, in relation to temperature inside the material.

face. Second, the amount of material at a given distance from the unit's center increases with the distance, particularly in cone-shaped piles. Accordingly, for conduction of a given quantity of heat, the temperature gradient  $\partial t/\partial x$  should decrease towards the surface. Since the two facts have opposite effects and the calculation does not have to be exact, we may assume a constant temperature gradient  $\Delta t/\Delta x$ . The situation is analogous for the oxygen concentration gradient  $\Delta c_v/\Delta x$  used below.

The flux of conducted heat (Q<sub>c</sub>) is

$$Q_c = -k \Delta t/\Delta x \qquad J/cm^2 s \tag{7}$$

For chips and sawdust, thermal conductivities k are not known, but other forest products give clues. Dry sawdust from a wood species of specific gravity 0.5 has the bulk density of  $0.5 \times \frac{1}{3} = 0.167$  g/cm<sup>3</sup>. For this density, 27 C, and 12% moisture content, thermal conductivities are for fiber insulating board 0.00036,

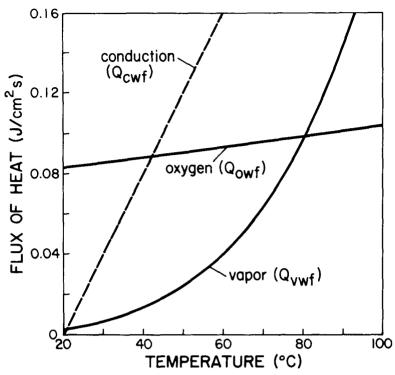


FIG. 2. Fluxes of heat produced by diffusing oxygen, conduction, and diffusing vapor in solid wood in fiber direction at 30% moisture content, in relation to temperature inside the material.

for particleboard 0.00044, for solid wood transverse to the fiber direction 0.000544, and for solid wood in the fiber direction 0.00104 J/cm s C (Kollmann and Côté 1968). Fiberboard and particleboard conduct heat essentially transversely, but because of discontinuities from piece to piece, their conductivities are lower than transversely in solid wood of the same density. Sawdust and chip piles have similar discontinuities. On the other hand, pieces in these piles are oriented at random, some with fibers pointing in the direction of heat flux. Considering these factors, the value for sawdust should average 0.0006 J/cm s C. In moist material, conductivity is considerably higher, being 0.001 J/cm s C near a 30% moisture content.

Thermal conductivities increase as temperature rises, particularly in porous moist wood (Kuhlmann 1962). In the center of self-heated piles the temperature generally hovers around 65 C. For the average temperature between this hot center and the surface, 43 C, thermal conductivity of sawdust may be 0.0011 J/cm s C. Walker and Harrison (1977) determined at 45 C the much higher value 0.003 J/cm s C. The difference is attributed to an unusually high moisture content of 138% in the sample of Walker and Harrison; water's thermal conductivity, 0.00562 J/cm s C, is after all much higher than that of dry wood.

For simplicity's sake, I shall use the 43 C value, 0.0011 J/cm s C, for all temperatures; at different degrees of heat the resulting error is small compared with the effect of temperature on transferring heat by diffusion of vapor, as discussed below. Conductivity of pulpwood chips, which are relatively coarse, should be

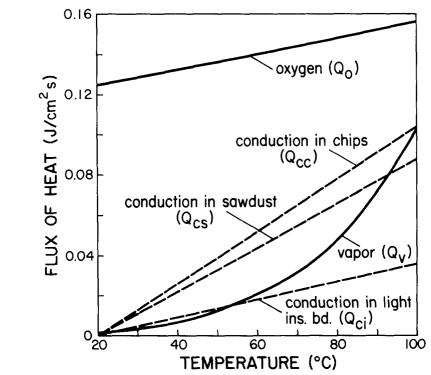


FIG. 3. Fluxes of heat produced by diffusing oxygen, conduction, and diffusing vapor in pulpwood chips, sawdust, and light insulating board at 30% moisture content, in relation to temperature inside the material.

somewhat higher than that of sawdust, around 0.0013 J/cm s C. Other conductivities of interest are listed in Table 1. Figures 1 to 3 include the corresponding fluxes  $Q_{\rm c}$  for the various forest products.

## HEAT TRANSFER BY VAPOR

Thermal conductivities increase so much with temperature in moist wood because wood moisture evaporates in warm zones only to condense in cool zones, delivering that vapor's latent heat. Moisture may evaporate inside a cell at the surface of the warm cell wall, diffuse across the cell cavity, condense on the

TABLE 1. Thermal conductivities of forest products at 43 C and 30% moisture content.

Product and direction	Bulk density (g/cm³)	Conductivity (J/cm s C)
Solid wood, transversely	0.50	0.00160
Solid wood, in fiber direction	0.50	0.00400
Light fiber insulating board, transversely	0.16	0.00045
Light hardboard, transversely	0.50	0.00080
Sawdust	0.17	0.00110
Pulpwood chips	0.19	0.00130

slightly cooler opposite wall of the same cell, migrate in bound form through the wall, and evaporate again.

Some of the evaporated moisture does not condense in wood, but rather diffuses all the way across the material into the surrounding atmosphere and carries its latent heat out of the wood. Heat transfer with temporary condensation and heat transfer without condensation are difficult to separate. The total of vapor-transferred heat reaches significant proportions at elevated temperatures. Vapor transfers so much heat at high temperatures because vapor pressure progressively increases as temperature rises, reaching atmospheric pressure at 100 C. In the following, heat transfer by vapor is assessed on the basis of the rate at which moisture migrates through forest products. Up to the frequent pile temperature of 65 C, the calculated flux  $Q_{\rm v}$  of heat in diffusing vapor may include some heat considered already in the flux  $Q_{\rm c}$  of conduction, but the error is insignificant.

Diffusion of vapor through voids of piles can be treated like diffusion of oxygen, using the diffusion coefficient  $D_v$  for vapor in air,  $D_v = 0.219(T/273)^{1.75}$  (Gray et al. 1972), for the average temperature between pile center and pile surface. Accordingly, the effective diffusivity would be  $0.16D_v$ . Measured moisture movement through fiber-saturated solid wood, transverse to fiber direction, corresponds to the effective diffusivity  $0.25D_v$  (Kubler 1957). Moisture migrates through moist wood much faster than does oxygen, because the cell walls that act as barriers for diffusing gas do conduct moisture in bound form from cell cavity to cell cavity. Though some wood moisture migrates as bound water and does not transfer latent heat as does vapor, this relatively fast movement in solid wood shows that in piles diffusion through pieces should be taken into account, besides diffusion in voids. Vapor migrates through piles containing voids probably as fast as moisture moves transversely through solid wood:

$$m_v = -0.25D_v \Delta c_v/\Delta x = -0.055(T/273)^{1.75} \Delta c_v/\Delta x$$
 g/cm<sup>2</sup> s (8)

For diffusion of vapor in solid wood in the transverse direction, I am using the same equation, though the amount of truly diffusing vapor is probably lower. In the fiber direction in solid wood (indexed wf), moisture moves roughly as fast as vapor in still air, but one-half of it may migrate as bound water without transferring latent heat, so that

$$m_{\text{vwf}} = -0.11(T/273)^{1.75} \Delta c_{\text{v}}/\Delta x$$
  $g/\text{cm}^2 \text{ s}$  (9)

In the calculation of the heat flux Q<sub>v</sub> of diffusing vapor,

$$Q_v = m_v L \qquad J/cm^2 s \tag{10}$$

the vapor concentration differential  $\Delta c_v$  (g/cm³) is chosen for 20 C and a relative humidity of 50% outside the material, with vapor-saturated hot air within the material. L stands for latent heat of vapor (J/g) as listed in tables of handbooks for the various temperatures. Calculated heat fluxes  $Q_v$  are shown in Figs. 1 to 3.

When air of 50% relative humidity surrounds wood, vapor diffuses out and cools the material even at ambient temperature. At slightly higher degrees of heat, flux of conducted heat  $Q_c$  by far exceeds the heat flux of diffusing vapor  $Q_v$ , but towards higher temperatures the migration of vapor rapidly increases, so that  $Q_v$  reaches  $Q_c$  according to Figs. 1 to 3 in light fiber insulating board near

54 C, in light hardboard at 80 C, in sawdust at 93 C, and in piles of chips at 100 C, whereas in solid wood flux  $Q_c$  seems to remain higher even at 100 C. Boiling the moisture in wood, however, raises the pressure above that of the atmosphere, so that vapor streams out rather than diffusing and flux  $Q_v$  dramatically increases. In wood that contains moisture, vapor transfers above the boiling point of wood's moisture in any case more heat than conduction does.

### DISCUSSION AND CONCLUSIONS

Temperatures in forest products rise until the total of flux of conducted heat  $(Q_c)$  and flux of diffusing vapor  $(Q_v)$  equal the flux of diffusing oxygen  $(Q_o)$ . According to Figs. 1 to 3, this stable state is reached in solid wood for transverse transport at 24 C, for transport in fiber direction at 39 C, in light hardboard at 27.5 C, in pulpwood chips at 87 C, in sawdust at 92 C, and in very light fiber insulating board near 100 C. In dry material, in which no vapor diffuses and thermal conductivity is lower, similar figures would show that the stable temperatures are about 28 C in solid wood transversely, 36 C in light hardboard, 55 C in solid wood in fiber direction, and far above 200 C in the permeable products. Of course, because of various assumptions and simplifications, these temperatures cannot be as accurate as their oddity implies; instead they give only the order of magnitude.

According to the figures, oxygen that diffuses into the relatively permeable materials—insulating board, sawdust, and chips—can raise temperatures to the boiling point of water, while in reality, in open piles of moist chips and sawdust, temperatures generally hover near 65 C and rise higher only after most of the moisture has evaporated (Springer personal communication). This discrepancy is explained by convection of hot air as in a chimney carrying large amounts of heat out of the piles. Since convection of air supplies oxygen as well as transferring heat, we must conclude that in open piles the concentration of oxygen remains high, rather than dropping to zero at a distance  $\Delta x$  cm below the surface, as assumed in the calculations.

The fact that larger piles of chips and sawdust self-heat more than do small piles (Riley 1979) confirms this conclusion. In the calculations, I assumed that diffusing oxygen can be consumed at any distance  $\Delta x$  cm from the surface. Consumption close to the surface is associated with steep oxygen concentration gradients, strong migration of oxygen, and corresponding high fluxes of heat Q<sub>0</sub>; but in this case heat losses by means of conduction and diffusing vapor are also high. Therefore, contrary to field observations, the calculated stable temperatures do not depend on pile size. Obviously, even in large piles not all diffusion oxygen is consumed, and the consumed amounts of oxygen as well as quantities of heat generated per unit pile volume are the same irrespective of pile size. Since heat generated deep in a large pile has a long way to go to reach the surface, a relatively high pile temperature is required to drive the heat out. Very likely, oxygen controls self-heating in open piles of chips and sawdust only at very high temperatures, where direct chemical oxidation approaches the rate of combustion. At moderate degrees of heat, pile temperature depends mainly on convection of air and can be controlled at temperatures up to at least 100 C by strong convection of cool air through the pile, as has been shown in another study (Kubler 1982b). Injected water holds temperatures down too, since hot water and steam carry

heat out. On the other hand, the water may block the path of cool air, so that pockets not reached by water self-heat even more than without injected water.

By contrast, in solid wood and in panels that lack voids between pieces, oxygen is consumed before it can diffuse to the center of the stack, and lack of oxygen limits generation of heat. This seems to be the reason why green lumber stacked without stickers does not self-heat noticeably; fungi rapidly spread and penetrate deep into each piece, but are forced to inactivity when they have consumed the "residence oxygen."

Lumber and relatively impermeable forest products self-heat in hot surroundings occasionally up to the point of ignition. At high ambient temperature, the products are usually bone-dry, and not cooled by diffusing vapor. Even so, diffusion of oxygen can be responsible only for small temperature increases, on the order of 6 C under side grain surfaces and about 20 C under end grain surfaces. We have to conclude that in this case hydrolysis and pyrolysis, as processes that do not consume oxygen, generate most of the heat.

In cases of self-heating in hot-stacked hardboard, or during tempering of hardboard, or in lumber in high-temperature kilns, it cannot be oxygen either that generates the bulk of heat inside these relatively impermeable products; again it must be hydrolysis and pyrolysis. Veneer and small wood particles in dryers are in intimate contact with oxygen and can theoretically react with oxygen. Whether they do react at significant rates compared with pyrolysis is another question.

#### REFERENCES

- Coulson, J. M., and J. F. Richardson. 1968. Chemical engineering, vol. 2, Unit operations. Pergamon Press, Oxford, United Kingdom. P. 111.
- Gray, D. E. (ED.), ET AL. 1972. American Institute of Physics handbook, 3rd ed. McGraw Hill, New York. Pp. 2-250.
- HAWLEY, L. F. 1952. Combustion of wood. Page 822 in L. E. Wise and E. C. Jahn, Wood chemistry, 2nd ed. Reinhold Publ. Corp., New York.
- KLASON, P., G. V. HEIDENSTAM, AND E. NORLIN. 1910. Investigation on dry distillation of wood. Zeitschr. f. Angew. Chemie 23(27):1252.
- KOLLMANN, F. P., AND W. A. Côté, Jr. 1968. Principles of wood science and technology, vol. 1, Solid wood. Springer-Verlag, Berlin, Germany. P. 249.
- KUBLER, H. 1957. Studies on movement of moisture through wood. Holz Roh- Werkst. 15(11):453-
- ——. 1982a. Heat release in thermally disintegrating wood. Wood Fiber 14(3):166–177.
- ----. 1982b. Air convection in self-heating piles of wood chips. Tappi 65(8):79-83.
- ----. 1982c. Self-heating of forest products due to residence oxygen. Wood Science 15(2):97-100.
- KUHLMANN, G. 1962. Study on thermal properties of wood and particleboard in relation to moisture and temperature in the hygroscopic range. Holz Roh- Werkst. 20(7):259–270.
- RILEY, R. G. 1979. Fires in raw material storage. Pulp Paper 53(14):100-103.
- WALKER, I. K., AND W. J. HARRISON. 1977. Self-heating of wet wood. New Zealand J. Sci. 20(2): 191–200.